A new high thermal-conductivity composite material for high-precision space optics

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ABSTRACT

This paper reports on the newly developed graphite-cyanate composite pipes for high-precision space optics such as the Solar-B optical telescope. Fundamental mechanical, thermal, and hygroscopic properties of unidirectional graphite-cyanate laminates were evaluated, first. The orientation of fibers in the pipe was designed to minimize longitudinal thermal deformation. Model pipes were fabricated based on the design, and have conducted a series of measurements to evaluate the thermal expansion behavior, the hygroscopic performance, the thermal conductivity, and the long-term stability. Excellent performance of the pipe was successfully verified and the material was found to be the most promising candidate for space optics structures.

KEY WORDS: composite material, graphite-cyanate, dimensional stability, thermal conductivity, material design

1. INTRODUCTION

The graphite-epoxy composites have been extensively used for space-borne optical instruments. It, however, has some known concerns; long-term dimensional stability (hygroscopic distortion) as well as low thermal conduction resulting in a significant temperature gradient in critical optical structures.

Recently, new pitch-based graphite fibers with high tensile modulus and high thermal conductivity have been developed and several space applications such as radiator panels\(^1\) and bus structures\(^2\) have been studied. Another important base materials of composites recently developed are cyanate resin systems. They have a lot of benefits for composites for space applications which include low outgassing, low moisture absorption, low microcracking, low dielectric constant and loss tangent. Space antenna, for example, composed of cyanate resins showed outstanding electrical performance and dimensional stability\(^3\). By combining these newly developed base materials, it is possible to fabricate a new composite material which has much higher thermal conductivity, and better hygroscopic performance as well as greater heat-resistance than conventional graphite-epoxy materials.
In this report, we evaluated fundamental properties of a graphite-cyanate composite, first, by fabricating unidirectional laminates. A new composite pipe for structures of space optics was designed, and model pipes were fabricated, and thermal stability, hygroscopic stability, and thermal conductivity was experimentally evaluated to verify material design.

2. MATERIAL PROPERTIES FOR PIPE DESIGN

2.1 Materials and fabrication of the laminates

Graphite fiber applied to the pipe was pitch based high modulus K13C fiber (Mitsubishi Chemical) which was chosen for its excellent thermal conductivity of more than 600 W/mK. Matrix resin to fabricate composites was EX1515 cyanate resin (Brite technologies) whose water absorption is much less than conventional epoxy resin systems.

Laminates were fabricated by laying-up prepreg sheets in which unidirectional K13C fibers impregnated with EX1515 resin. Zero degree prepreg sheets were laid up, vacuum bagged and autoclave cured for 4 hours at 170 °C and 1.3 MPa pressure. The volume fraction of the laminates were approximately 60%.

The laminates were thermal cycled prior to the test for pre-conditioning. This is to evaluate practical properties of materials in orbit because some mechanical properties of plastic matrix composites degrade due to appearance of microcracking in the matrix resin after thermal cycled. The upper and lower limit of the temperature of the thermal cycle was ±150°C respectively. The number of cycle was 20.

2.2 Mechanical properties

Tensile moduli of the laminates were evaluated both in the direction of 0 degree and 90 degree to the fiber at room temperature. The moduli were obtained based on the technique in ASTM D3039 with an Instron type testing machine IS-10T (Shimadzu). 20 plies of prepreg sheets were laid up. The specimen was straight-sided and of constant cross section of 25 mm x 2 mm The length of the specimen was 250 mm. Glass fiber reinforced plastic tabs were bonded to the specimen. To determine Young's modulus and Poisson's ratio, 0/90 degree cross strain gages were attached to the specimen. Crosshead speed was 2 mm/min.

Tensile tests were also performed in the direction of 45 degree to evaluate shearing modulus. When the tensile moduli, \( E \), in the direction of 0, 45, and 90 degree and Poisson's ratio, \( \nu \), are obtained, the shearing modulus, \( G \), is calculated as:

\[
\frac{1}{G} = \frac{1}{E_{45}} - \left( \frac{1}{E_0} + \frac{1}{E_{90}} \cdot \frac{2\nu_0}{E_0} \right)
\]  

(1)

Five specimens were tested in each direction. Average tensile Young's modulus, shearing modulus, and Poisson's ratio are listed in Table 1. Very high modulus in the fiber direction (almost 3 times as much as steel) and strong anisotropy of this

<table>
<thead>
<tr>
<th>Young's modulus</th>
<th>Shearing Modulus</th>
<th>Poisson's ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 degree GPa</td>
<td>90 degree GPa</td>
<td>GPa</td>
</tr>
<tr>
<td>592</td>
<td>5.29</td>
<td>4.95</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.28</td>
</tr>
</tbody>
</table>

Table 1 Mechanical properties of unidirectional laminates
material were shown.

2.3 Thermal expansion

A thermal expansion analyzer DL-7000y-RH (Shinku-Riko) was employed to determine coefficients of thermal expansion (CTEs) of the laminates. Thirty plies of the laminates were cut into specimens of 3.6 mm wide, 65 mm long and 3.0 mm thick in the direction of both 0 degree and 90 degree. Heat-up rate in the test was 5°C/min.

Five specimens were measured. Typical CTE values of the laminates are plotted in Fig. 1. Average values at around room temperature were -1.3 ppm/°C in the direction of 0 degree and 37.7 ppm/°C in the direction of 90 degree, respectively.

![Graph](image1.png)

(a) 0 degree.

![Graph](image2.png)

(b) 90 degree

Fig.1 Coefficient of thermal expansion of unidirectional laminates.
2.4 Moisture absorption

Deformation due to moisture absorption was evaluated by measuring coefficients of moisture expansion (CMEs) and total amount of moisture absorption. CMEs of the laminates were measured with a thermo-mechanical analyzer TM7000 (Shinku-riko). Figure 2 shows the specimen. 20 plies of The laminates were cut into specimens of 4.0 mm wide, 100 mm long and 2.0 mm thick in the direction of both 0 degree and 90 degree. Aluminum foil was attached on both side of the specimen.

![Diagram of specimen](image)

Fig. 2 Specimen for moisture absorption testing.

Five specimens were dried up in an oven at 100 °C for 40 hours prior to the test. Longitudinal deformation, \( \Delta L \) and weight gain, \( \Delta w \) were measured after 24 hours in the chamber where the temperature was 30 °C and the relative humidity was 70%. The value of CME, \( \beta \) was calculated as:

\[
\beta = \frac{\Delta L}{L} / \frac{\Delta w}{w}
\]

(2)

The specimens were then kept in another chamber in the same temperature and humidity condition as CME measurement for more 20 days to evaluate weight gain.

Table 2 summarizes the moisture absorption properties. Total amount of moisture absorption of the material was found about one order less than that of conventional epoxy resin matrix composites. Little hygroscopic deformation was predicted especially in the fiber direction.

<table>
<thead>
<tr>
<th>Table 2 Hygroscopic properties of unidirectional laminates</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coefficient of moisture expansion</td>
</tr>
<tr>
<td>0 degree</td>
</tr>
<tr>
<td>%/%</td>
</tr>
<tr>
<td>-0.01</td>
</tr>
</tbody>
</table>
2.5 Thermal conductivity

Thermal conductivity of the material was evaluated by means of laser-flash technique. Thermal constant analyzer LF/TCM FA8510B (Rigaku electronic) was employed. The result was shown in Table 3. Thermal conductivity in the fiber direction was more than 300 W/mK which is twice as much as that of general aluminum alloys for space structures.

<table>
<thead>
<tr>
<th></th>
<th>0 degree</th>
<th>90 degree</th>
</tr>
</thead>
<tbody>
<tr>
<td>W/mK</td>
<td>335</td>
<td>1.5</td>
</tr>
</tbody>
</table>

3. DESIGN AND FABRICATION OF MODEL PIPES

3.1 Material design

Based on the evaluated properties of the uni-directional laminates, fiber orientation in the pipe was determined. The pipe applied to the structure of high-precision space optics requires to be stable in length in severe thermal conditions. Thermal expansion of the pipe, therefore, should be designed to be zero in longitudinal direction. The CTE of the pipe was predicted based on a lamination theory, and several types of stacking sequence were evaluated to minimize the value.

In addition, thermal conductivity of the pipe should be high to avoid thermal gradient in the pipe due to the concentration of heat in such case as the focusing of the light ray. Another consideration in the orientation design was formability of the pipe. Because of high stiffness of the graphite fiber applied here, bias angle of the fiber is limited. By these considerations, a lay-up of the pipe was determined as \( [(0)_5/(45)_{(-45)}/(60)_{(-60)}/(0)_{(70)}/(70)_{(-70)}] \).

3.2 Fabrication of the model pipe

Model pipes whose length were 200 to 600 mm were fabricated to verify material design. Figure 3 shows the configuration of the model pipe. The pipes were fabricated by sheet winding method. In the method, prepreg sheets were cut and piled according to the designed orientation. Then, a mandrel was wound by the laminates and shrinking tape. After cured in an oven at 170 °C for 4 hours, the mandrel was pulled off. Because the outer layer is sometimes peeled off along the unidirectional fiber in this fabrication process, two layers of cloth were laid on both inside and outside of the pipe. Figure 4 is a photograph of the model pipe.

4. EVALUATION OF MODEL PIPES

4.1 Thermal deformation
K13A/EX1515 cross ply laminate, 2-ply ($0^\circ$/$90^\circ$)

K13C/EX1515 unidirectional ply laminate
Stacking sequence:
$(0)_6/(+45)(-45))_4/((+60)(-60))_6/(0)_2/((+70)(-70))_4$

cross sectional view of the pipe

Figure 3 Lay-up of composite pipes.

Fig. 4 A model pipe.
Figure 5 shows the experimental set-up to evaluate thermal deformation of the pipe. The pipe on which 8 thermocouples were attached was wound with tape heater and wrapped with glass wool blanket for thermal insulation. One end was fixed to the angle plate through a ultra low elongation (ULE) glass plate for insulation. A reflecting mirror was bonded to another side, through the optical adapter.

Thermal dependent displacement was measured by laser interference method. A Helium-Neon laser beam whose wave length is 0.316 μm is split by a beam splitter. One beam is reflected off the specular surface on the mirror attached to the pipe. The reflected beam and another splitted beam interfere. By counting the times of change in intensity of the beam, the displacement is evaluated. Accuracy of this method was verified by evaluating a reference specimen made of copper alloy.

Two pipes of 600 mm length were evaluated in the temperature range from 26 °C up to 52 °C both in heating and cooling process. The result is shown in Fig. 6. The displacement changed linearly dependent on the temperature. By subtracting deformation of the optical adapter made of stainless steel from the measured displacement, the CTE of the pipe was calculated as -0.8 ppm/°C, which is almost same as predicted value of -0.6 ppm/°C. Difference of these values are mainly due to the effect of the surface layer composed of K13A graphite fiber cloth. By taking the surface layer into account and
slightly increasing volume fraction of matrix resin, more precise control of CTE is possible.

Since the measured CTE matches well with the predicted value, we will be able to design and fabricate the pipes with much lower absolute value of CTE (<0.1 ppm/°C required for the zero-CTE structural pipes for Solar-B optical telescope).

4.2 Hygroscopic deformation

Long term stability of the pipe was evaluated by measuring weight gain and change in length of the pipe. First, the pipes were kept in the oven at 100 °C for 40 hours to be dried up. The weight was measured by an electrical balance, and the length was measured by a three dimensional length measuring machine. Then the pipes were kept in an environmental chamber where the temperature was 60 °C and the relative humidity was 95%. After 7 days, the weight and the length were measured again.

Three pipes were evaluated, and the results are summarized in Table 4. Judging from the weight gain, moisture absorption of the pipe was almost saturated. Average value of deformation is 0.5 μm, which is less than the accuracy of the

Table 4 Hygroscopic properties of model pipes.

<table>
<thead>
<tr>
<th>Moisture expansion</th>
<th>Moisture absorption</th>
</tr>
</thead>
<tbody>
<tr>
<td>Displacement</td>
<td>Expansion</td>
</tr>
<tr>
<td>mm</td>
<td>ppm</td>
</tr>
<tr>
<td>-0.0005</td>
<td>2.5</td>
</tr>
</tbody>
</table>
measuring machine. Compared with the predicted value of 0.6 μm based on a lamination theory, the result is considered to be appropriate. Longitudinal deformation due to moisture absorption of the developed pipe, therefore, is at most 3 ppm, which is much less than that of conventional epoxy matrix composites (about 70 to 160 ppm).

4.3 Thermal conductivity

Figure 7 shows the experimental set-up to evaluate thermal conductivity of the pipe. The pipe with 8 thermo-couples was folded by an insulator and set on grass fiber reinforced plastic stands in the vacuum bagged chamber. A sheet heater was attached around one edge of the pipe. Time dependent change in temperature of each measuring point was monitored. When the heat input to the pipe is \( Q \) (W), thermal gradient at the point \( \Delta L \) (m) apart from the edge is \( \Delta T/\Delta x \), and time dependent change in the temperatures \( \Delta T/\Delta t \), thermal conductivity of the pipe, \( \lambda \), is calculated as:

\[
\lambda = \frac{Q - S \left( \frac{\Delta L}{2} \right) c \rho \left( \frac{\Delta T}{\Delta x} \right)}{S \left( \frac{\Delta T}{\Delta x} \right)}
\]

where, \( S \): cross sectional area, \( c \): specific heat, \( \rho \): density.

The measured thermal conductivity was 111 W/mK which is a little less than the predicted value of 140 W/mK. This is because the anisotropy of this material reduces circumferential thermal diffusion. The evaluated value, however, was one or two order higher than conventional graphite fiber composites and excellent thermal performance of the developed pipe was verified.

![Diagram of experimental setup](image)

**Fig. 7** Experimental set-up to evaluate thermal conductivity of pipes.
5. CONCLUSIONS

In this paper, a new composite pipe for the structure of space optics was newly designed by introducing a high performance pitch based graphite fiber and a high moisture resistant resin. The results are summarized as follows;

(1) Mechanical properties, coefficients of thermal expansion, moisture absorption, and thermal conductivity were experimentally evaluated by fabricating unidirectional laminates.

(2) Based on the properties, the fiber orientation of the pipe was designed to minimize longitudinal thermal deformation.

(3) Model pipes were fabricated, and deformation in thermal and hygroscopic environment and thermal conductivity were evaluated.

(4) Excellent thermal and hygroscopic stability and high thermal conductivity were verified, and the developed pipe was found to be the most promising candidate for space optics structures.

(5) The demonstrated performance allows us to proceed to the design and the fabrication of the zero-CTE pipes for Solar-B optical telescope.

6. ACKNOWLEDGEMENTS

This research was supported by a Grant-in-Aid for Scientific Research of the Ministry of Education, Science, Sports, and Culture in Japan (#07554047 awarded to S. Tsuneta).

7. REFERENCES